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TITLE LIQUID CENTRIFUGATION FOR NUCLEAR WASTE TARGETING

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Liquid Centrifugation for Nuclear Waste Partitioning

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Abstract

The performance of liquid centrifugation for nuclear waste partitioning is examined for the Accelerator Transmutation of Waste Program currently under study at the Los Alamos National Laboratory. Centrifugation might have application for the separation of the $LiF \cdot BeF_2$ carrier salt from heavier radioactive materials fission product and actinides, in the separation of fission product from actinides, in the isotope separation of fission-product cesium before transmutation of the ^{137}Cs and ^{135}Cs, and in the removal of spallation product from the liquid lead target. It is found that useful chemical separations should be possible using existing materials for the centrifuge construction for all four cases with the actinide fraction in fission product perhaps as low as 1 part in 10^7 and the fraction of ^{137}Cs in in ^{133}Cs being as low as a few parts in 10^5. A centrifuge cascade has the advantage that it can be assembled and operated as a completely closed system without a waste stream except that associated with maintenance or replacement of centrifuge components.

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I. Introduction

The Los Alamos National Laboratory is examining possibilities for the transmutation of long-lived radioactive wastes to short-lived or stable forms by neutron-induced transmutation using neutrons produced by a powerful accelerator[1]. This Accelerator Transmutation of Waste program (ATW) is a system consisting of five major components; the input chemistry, the accelerator, the liquid lead target for the proton beam which produces neutrons, a surrounding blanket where the transmutation takes place and where fission energy is generated, and the output chemistry. Two commercial applications of

this technology are under study including the transmutation of waste from commercial nuclear power plants, and the generation of fission energy with concurrent transmutation of the radioactive wastes. In both cases the long-term radioactivity of the remnant waste would be essentially eliminated. These systems must compete in the marketplace with other sources of electric power While the accelerator, target, and blanket offer very interesting design challenges, no technical barriers to the implementation of this technology have been encountered in the study of those systems to date. However the input and output chemistry do offer significant technical challenges in terms of the choice of chemistry, the effectiveness of the chemistry to provide separations to high purity, the avoidance of waste streams, and cost competitiveness. The economic competitiveness of the systems may depend to a large degree on the chemistry applied.

The interplay between the chemistry and the other components of the system is very strong. It is tempting to select a well developed and well understood chemistry which will then dictate the size of the accelerator and important features of the target and blanket. For example, in systems where the residence time of the waste in the transmuter is short, the waste and fuel probably must be in the form of liquids. The selection of aqueous chemistry will almost certainly require continuous transport of the waste and nuclear fuel in the transmutation system in an aqueous medium. The containment of the water at high temperatures causes an incompatibility between efficient heat removal with effective conversion to electric power and low parasitic neutron capture, which is required for optimal neutron economy and the smallest possible accelerator.

Arguments can be made based on the neutron economy considerations of Ref. 1 that an aqueous blanket will exhibit doubtful economic competitiveness when it addresses both the problem of burn-up of the 7.5% of the fission products which are long-lived and the transmutation of all of the plutonium and higher actinide waste. This conclusion derives from the physics principles of neutron production by spallation, neutron liberation in fission, the slowing-down and diffusion of neutrons, transmutation nuclear data, heat transfer, and the nuclear and performance properties of structural materials. Rather than allowing aqueous radiochemistry to impose neutronic performance limitations on the target-blanket system, it is of interest to design the target-blanket for optimal performance and then to see what demands this optimal neutronic performance will place on the chemistry.

The Los Alamos studies directed along the lines of optimal neutronic performance are focusing onto a graphite-moderated helium-gas-cooled system with the actinide and fission product being carried as fluorides in LiF-BeF_2 molten salt[2]. The optimal performance of this system requires that the fission product be continuously removed from the carrier salt. The chemistry demands are severe requiring either pyrochemistry with some undeveloped and perhaps difficult steps involved or the transformation to aqueous chemistry and back again continuously to allow continuous reprocessing. For this reason the present work is based on physical chemistry methods which do not rely on

the chemical properties of the elements but simply on the large difference in mass and density of the carrier salt, the fission products, and the actinides. The molecular masses of these three classes of these fluorinated materials are approximately 38, 157, and 350 suggesting effective separation by centrifugation.

While most of the long-lived fission products can be dealt with effectively via transmutation, the nuclides ^{135}Cs and ^{137}Cs require isotopic separation before transmutation can be effectively applied. This can be accomplished implicitly for the energy generating system with concurrent waste transmutation[1]. However, for commercial waste transmutation this option is not available and isotope separation for Cs appears necessary; it might not be essential for Sr since the neutron penalty is much less severe for Sr than for Cs. A second objective for this study therefore is to evaluate the prospects of centrifugation for Cs isotope separation.

A third objective is to evaluate the removal of spallation product from the liquid lead target by centrifugation in order to avoid chemistry processes. The spallation products can be divided rather conveniently into groups with densities either substantially lower or higher than the lead density. This density grouping is the basis for centrifugal cleansing of the lead target

II. Derivation of the Separation Equation

All previous centrifugation activity in nuclear technology has been applied to uranium enrichment and to the enhancement of reaction rates by centrifugal contactors.[3-5] The separation of materials based on their difference in density is a very well developed technique for the case of bio-medical research[3]. One well developed method called the sedimentation-equilibrium method relies on the establishment of a density gradient in an aqueous salt solution by centrifugation. If the solution also contains one or more proteins at low molar fraction, a protein will drift under the centrifugal field to a position in the solution where its density is matched by the solution. This approach and variations of it are common practice in modern biological laboratories. Most such university laboratories have at least one centrifuge capable of creating forces 10^5 times that of gravity; these centrifuges can be acquired at the cost of perhaps $20,000. The physical basis for these centrifuges therefore is well established, and there is no question that the basic concept is sound. However the review texts on centrifugation do not include the case of interest here. The fundamental equations are presented in them however so that a starting point is available. The problem is to find the appropriate simplifications to allow useful analytical solutions to be developed which illustrate the physics principles involved. The imposition of appropriate boundary conditions and the estimates of sedimentation velocities also are necessary. We derive the fundamental equation below.

A number of assumptions are made in the derivation which are commented on briefly. The derivation is done for a binary system. While the extension to more than two components is in principle straightforward, the

actual solutions are expected to be complicated and not very useful for illustration of the physical principles. In actual practice numerical solutions probably will be required of systems with more components. Incompressibility of the liquids is also assumed. This is not strictly correct since at the high pressures involved the materials do compress by a few volume percent. Since the energy associated with this compression is small compared to that required for the separations, the compressibility is neglected. We assume ideal liquids; that is that the intermolecular forces between molecule A-A, B-B, and A-B are all the same. We show near the end of this report that the relaxation of this assumption does not significantly affect the centrifugation performance. We assume that the partial molar volume is the same as the molar volume. The difference between the molar volume and the partial molar volume is assumed to be the least when the materials are similar; the materials in the present case all are fluoride salts. The calculations address equilibrium rather than the dynamics of the approach to equilibrium. The rate of approach to equilibrium are however estimated separately.

The Gibbs free energy for the system depends on the radius, the pressure, and the molar composition of the mixture. At equilibrium the change in free energy is zero so that we may write the Gibbs free energy per mole G_i for the component i as

$$dG_i = (dG_i/dr)dr + (dG_i/dP)dP + (dG_i/dx_i)dx_i = 0 \qquad (1)$$

where P is the pressure which depends on the radius, x_i is the molar concentration of component i, and the parenthesis indicate partial derivatives. The term $(dG_i/dr) = -Force_{centrifugal} = -M_i r\omega^2$. For a compressible fluid such as a perfect gas, $(dG_i/dP) = 1/P$ and this is the term which is used for gas separations. For an incompressible fluid[6], $(dG_i/dP) = V_i$ where V_i is the partial molar volume of component i. The pressure varies with r according to the relationship $dP = \rho\omega^2 r$ where ρ is the mixture density. Finally we have the Gibbs free energy for component i as $G_i = G_i^o(T,P) + RT\ln(\gamma x_i)$ where R is the gas constant, T is the temperature and γ is the activity coefficient which is unity for an ideal liquid. Inserting the above definitions into 1 gives

$$-M_i\omega^2 r dr + \rho V_i \omega^2 r dr + RT dx_i/x_i = 0 \qquad (2)$$

for the component i, which is also given on page 135 of Ref. 3. The density is not constant with radius since the mole fraction of each material changes with r in response to the change in centrifugal force with radius and the components of the liquid in general have different densities. Combining terms in 2 and assuming a binary system for which $x_a + x_b = 1$, we have

$$(\rho V_a/M_a - 1)\omega^2 M_a r dr/RT + dx_a/xa = 0 \qquad (3)$$

To proceed we must write an expression for ρ. In an unspecified volume let us assume that there are x_a moles of light material and x_b moles of heavier

material. The total mass in this volume is $x_aM_a + x_bM_b$. The total volume of this material is $x_aV_a + x_bV_b$ where we assume that the molar volume and the partial molar volume are the same. The density ρ is

$$\rho = (x_aM_a + x_bM_b)/(x_aV_a + x_bV_b) = [x_a(M_a - M_b) + M_b]/[x_a(V_a - V_b) + V_b] \quad (4)$$

Substituting 4 in 3 and using $x_a + x_b = 1$, we may write 3 with the notation $x = x_a$ as

$$d(r^2)[M_a - (V_a/V_b)M_b]\omega^2/2RT = (dx/x)[1 - x(Vb - Va)/Vb]/(1 - x) \quad (5)$$

For the centrifugation of interest here, a boundary condition is required which conserves volume in the centrifuge. If we fill the centrifuge with a solution with a molar concentration of the lighter component of x_0, the heavier material will move to the outer radius and the lighter material to the inside. However under the assumptions of (1) incompressibility and (2) that the partial molar volume is the same as the molar volume, the total volume of x_0 in the system must remain unchanged. Consider then a cylindrical rotating volume of inner radius r_1 and outer radius r_2 and a height h containing x_0 moles of the lighter component. The volume occupied by the lighter material is $\pi(r_2^2 - r_1^2)h[x_0V_a/(x_0V_a + (1-x)V_b)]$. The equilibrium concentration of x varies with r but the volume of lighter material remains the same. Therefore conserving the volume of the lighter material we have

$$2\pi hrdrxV_a/[xV_a + (1-x)V_b] = h\pi(r_2^2 - r_1^2)x_0V_a/[x_0V_a + (1-x_0)V_b] \quad (6)$$

Rewriting 5 as

$$x = (dx/dr)(1/r)\{[1 - x(V_b - V_a)/V_b]/[1 - x]\}\{RT/\omega^2[M_a - (V_a/V_b)]\} \quad (7)$$

and using 7 in 6 yields upon performing the integral

$$[1 - x(r_1)/(1 - x(r_2)] = \exp(M_aV_b/V_a - M_b)\omega^2(r_2^2 - r_1^2)/(2RT[1 + (-1 + 1/x_0)V_b/V_a] \quad (8)$$

Equation 8 is the fundamental expression for calculating the centrifuge performance in this report. To gain some insight into its use, it is helpful to consider the case of isotopic separation in a liquid for which $V_a = V_b$. Equation 8 then becomes

$$[1 - x(r_1)/(1 - x(r_2)] = \exp(M_a - M_b)\omega^2(r_2^2 - r_1^2)x_0/2RT \quad (9)$$

It shows the same interesting result as for centrifugation of gases[3,4] in that the separation depends on the difference of the molecular weight and not $\Delta M/M$. Therefore heavy materials are separated as readily as light materials and for all molecular forms the equilibrium separations are the same. Of course the density of the material in the centrifuge is very different for a gas and an incompressible liquid, the amount of material in the liquid is many times that in

the gas for the same volume. On the other hand the viscosity of the gas is much lower than that of the liquid so that the separation rates are greater for the gas and therefore the volume throughput is higher for the gas centrifuge. Whether the mole processing rate is higher for the gas depends on a detailed analysis of the two systems. We use expression 9 later to consider the isotopic separation of Cs.

Returning to equation 8 we see that the leading term in the exponential can be written $V_b(M_a/V_a - M_b/V_b) = V_b(\rho_a - \rho_b)$ so that no separation is possible if the densities of the two materials are the same. The equilibrium separation of incompressible liquids then depends ultimately on the difference in densities, on the ratio of molar volumes, the molar volume of the heavier component, and on the initial molar fraction of the lighter material x_0.

III. Centrifuge Parameters

The form of the centrifuge which will be considered here is that of a uniform cylinder as shown in Fig.1. This unit is basically a rotating pipe with a central core and the capability to extract and feed two product streams at different radii. The capability to feed two streams is not considered here and so both inputs feed the same material. Table 1 shows the performance of several types of materials suitable for centrifuge construction[4]. Most of the materials operate with tangential speeds of 400 to 500 m/s although advanced materials can operate at about 700 m/s. For the studies here we assume a speed of 425 m/s and a speed for an advanced system of 700 m/s. The inner radius is taken to be 5 cm; the outer radius of 15 cm gives rotation speeds of 451 and 743 rps respectively. Most turbomolecular pumps operate at peripheral speeds of about 400 to 450 m/s with ball bearings.

At the bottom of the Table 1 the conditions for the various resonances are shown. The resonances are important since operation at resonance for any substantial period might damage the centrifuge. Therefore if the centrifuge is designed to operate above the first resonance, it must be built with powerful drive motors and brakes in order to move the centrifuge through the resonance quickly. We see that for these systems the first resonance appears at a length-to-radius ratio of about 14. This means that a device with a radius of 15 cm can be about 2 meters long without concern for resonance problems. The maximum volume is then about $\pi(15^2-5^2)200 = 125$ liters. The radiation levels around the centrifuges will be exceptionally high and the temperatures will also be high. These conditions might be a problem for the bearings and their lubrication. Therefore it would be highly desirable if the centrifuge could be built with magnetic suspension bearings (as is the case for the LANSCE chopper which runs at 600 rps) or air bearings. In either case these types of bearings probably will require a vertical orientation of the centrifuge.

IV. Separation of Actinides and Fission Products

The transmutation of actinide is by fission and the fission products must be removed continuously in order not to place a burden on the neutron economy and to allow the separation of the long-lived fission products for transmutation also[1]. For the molten salt based system, the actinides and fission products are present as a small molar fraction in a carrier salt consisting of a LiF-BeF_2 mixture. The first step in the required separations is the removal of most of the light carrier salt from the heavier actinide and fission product fluorides and the return of this carrier salt to the transmuter. The second step is the separation of the fission product from the actinide so that the actinide can be returned to the transmuter and so that the fission product can be further processed for separation of the long-lived constituents which must be transmuted to stable or short lived material. For this second step removing the fission product completely from the actinide is not required since the actinide will be returned to the transmuter. However, the actinide should be removed "completely" from the fission product so that there is "no" contamination of the actinides in the stable or short lived fission products which do not need to be transmuted.

For a transmutation system using molten salt as the carrier of actinide material, a successful centrifuge-based separation system would offer the enormous advantage that the fission product and actinide can be continuously separated in a closed system without a waste stream. That is, a physical rather than a chemical method is implemented so that the additions of reagents, solvents, etc. is not required, and therefore any waste streams associated with these chemicals are eliminated.

The first step, which is the carrier salt separation, does not require highly pure separations for either the carrier or the heavier materials since the salt is fed directly back into the blanket and the carrier separation must be followed by the fission product-actinide centrifugal separation. The carrier separation is made difficult by the fact that the heavier fraction is a rather small fraction of the total salt volume. We begin the performance evaluation with a listing of the relevant parameters.

Volume of carrier salt in the blanket	2800 liters
BeF_2-LiF mole ratio	16/71.7
BeF_2 density	1.99
LiF density	2.64
Molecular Weight of BeF_2	47
Molecular Weight of LiF	26
Density of carrier salt	2.52
Molecular weight of carrier salt	38.3
Molar volume of carrier salt	15.2 cm^3
Moles of carrier salt in the blanket	180,000
Actinide inventory in the blanket	280 Kg
Actinide concentration in the carrier salt	100g/liter
Actinide fluoride density	6.7
Molecular weight of actinide fluoride	351
Molar volume of actinide salt	52 cm^3

Mole concentration of actinide	0.65 %
Fission product fluoride density	4.0
Fission product fluonde molecular weight	157
Mole fraction of fission product in the carner	0.74 %
Mole fraction of fission product plus actinide	1.39 %
Density of actinide plus fission product	5.26
Molecular weight of actinide plus fission product	248
Molar volume of actinide plus fission product	47
ω	2827/s, 4397/s
T	873 K (600 °C)

The mole concentration of fission product in the salt depends on the choices for up-front processing and on the residence time for the fission product feed. We assume that no separation of the waste is done in the up-front processing except the removal of the volatile fluorides uranium and zirconium. All of the waste has been fluorinated[7] and is fed into the molten salt of the transmuter. We assume further that the system is transmuting the fission product waste from two commercial power reactors operating at 3000 MWt which produce 1000 Kg/ year of fission product waste. The system burns the waste from these and also produces a total of 1500 MWt of fission power so that the total amount of fission product which passes through the system is 2500 kg/year. The carrier salt is processed continuously at a rate such that the average residence time of a fission product atom in the system is about 30 days. The amount in the molten salt at any time is 2500X30/360 = 208 kg. Since the average molecular weight of the fission product is 0.157 kg, there are 208/0.157 = 1324 moles of fission product in the system. The fraction of fission product in the carrier is then 1324/180000 = .74 %. Therefore the mole fraction of fission product plus actinide in the system is 0.65% + 0.74% = 1.39 . These fractions may be used to calculate the molar volumes of carrier and the "heavies" and the average molecular weights of these same mixtures. For use in δ we then have the parameters

$V_a = 15.2\ cm^3$
$V_b = 47\ cm^3$
$M_a = 38.3$
$M_b = 248$
$x_0 = 1 - .0139 = 0.986$
$r_1 = 5$ cm
$r_2 = 15$ cm
$\omega = 2827, 4397$
$R = 8.31$ joules /K
$T = 873$ K

The mole fraction of the carrier salt starts out at 0.986 and in the first stage of centrifugation it is separated to C.994 at r_1 and 0.978 at r_2. The enriched carrier is then returned to the blanket and the material collected at r_2 is sent on to the second stage for further removal of carrier. The material from the first stage divides into two streams of equal flow rate so that half of the material

is sent on to the second stage. Fig 2 shows how the concentration of heavies changes for the same centrifuge parameters as the material flows through successive stages. Note that 8 stages are required to reach a molar fraction of heavies of 0.65 for $\omega = 2827/s$. This corresponds to a volume fraction of carrier of 0.38. Note that for the higher value for $\omega = 4397$, the same separation can be achieved in five stages since the centrifugal force is higher by about 2.5. With this higher field the drift rates are also faster by a factor of 2.5.

An assembly of centrifuges are shown in Fig. 3 with a possible concept for interconnection. The material from the first centrifuge at .985 concentration is sent into the second and emerges on the outside at a concentration of 0.965 and goes on to the third centrifuge. The material emerging from the inside has a concentration of .991 and is returned to the blanket. The concentration from the inside of the third centrifuge is 0.985. This is slightly better than the feed to the first centrifuge, so it is sent back to the first rather than into the blanket and recycling begins. It can be seen from Fig. 3 that in all successive stages the concentration from the inner radius is more favorable than the material in the blanket and that it can be reentered into the centrifuge assembly two stages back.

There are two important results from this recycling. The first is that after the second stage all material stays in the assembly meaning that 1/4 of that which enters emerges at the eighth stage with a molar concentration of 0.658. The second is that because of the recycling, the concentration of the output material is somewhat better than 0.658 because at every stage of recycle the recycled material from a downstream stage is more concentrated in "heavies" than the material coming from the upstream stage. The size of this effect has not been carefully analyzed but is estimated to improve the output molar concentration from .658 to .621. A final point that should be made is that the improvement in the heavies concentration is greater the greater the input heavies concentration. If there is actually a larger heavies concentration in the blanket to begin with than that chosen here, the effectiveness of the same centrifuge assembly would be very substantially enhanced. The concentration assumed of 0.0139 is conservative and the concentration in the real system might be a factor of two or three higher.

The centrifugation system shown in Fig. 3 reduces the carrier concentration from 0.986 to about 0.621. The heavies concentration has been increased from .0139 to 0.379 or by a factor of 27. These figures are equivalent to the separation and return of 97.7 % of the carrier to the blanket. It should also be noted that while the molar concentration of the carrier is 0.621, the volume fraction of the carrier is 0.375. Having eliminated nearly all of the carrier salt, the next step is to separate the actinide from the fission product in a subsequent centrifugation arrangement. To simplify the problem so that the binary equation can be applied, we neglect the remaining carrier salt. In the real situation it will come out of the subsequent centrifuge system mixed with the fission product. The actual composition of the fission product-actinide mixture is uncertain but the components are approximately equal and we assume equal molar quantities of each. The parameters for entry into 8 for the actinides as heavy

component b with assumed composition MF_6 and the fission product as the lighter component a with assumed composition MF_2 are as follows:

M_a = 157 grams
M_b = 350 grams
V_a = 39.25 cm^3
V_b = 52 cm^3
x_0 = 0.5
ω = 2827/s, 4397/s.

The purpose is to separate the fission product from the actinide so that the fission products which must be transmuted can be partitioned from the stable and short-lived fission products. The actinide fraction is returned to the blanket. The actinide fraction can contain a significant amount of fission product since the actinide is to be returned directly to the blanket. However it is important to have very little contamination of the fission product with the actinide in order to avoid further chemical processing to separate the several actinide elements from the fission product.

The performance of a chain of centrifuges for the two angular speeds are shown in Fig. 4 where the fraction of the actinide in the fission product is shown as a function of the number of stages assuming no feedback. The objective for the purity of the fission product is somewhat uncertain, but it is seen that a stream of 0.9999 purity could be obtained with ten centrifuges at the slower speed. At the higher speed, this purity level could be exceeded in four stages and 0.9999999 purity could be reached in six stages. Clearly there is very high leverage in increasing the speed as much as practical. Since the volume of the fission product -actinide mixture is smaller by about a factor of 1/0.139 = 70 than the carrier salt, much smaller throughput rates are required for the actinide-fission product separation and the actinide-fission product separations centrifuges could be much smaller and fewer in number. Higher centrifugal fields can be more readily achieved in smaller volumes so that very high quality separations might be achieved for the fission product-actinide separation.

In summary, we have described two set of centrifuges with the first set feeding the second. The first set removes the carrier salt returning it to the blanket and the second set cleanses the actinide from the fission product returning the actinide to the blanket for eventual transmutation by fission. The two set of centrifuges along with the blanket constitute a closed system with the only outlet being the purified fission product. There is no requirement for solvents, extractants, etc. and the associated problems of dealing with mixed hazardous waste. These prospective advantages are sufficiently notable that it is worth considering how the centrifuge concept might perform for two other applications. The first is isotope separation which is necessary for neutron-economic transmutation of ^{137}Cs. The second is the removal of spallation product from the liquid lead target. We examine the prospects for these next.

V. Isotopic Separation of ^{133}Cs from ^{137}Cs

If the transmutation of ^{137}Cs is attempted without isotope separation nearly all of the ^{133}Cs must be transmuted by successive neutron capture to ^{136}Cs or ^{137}Cs before it could be converted to stable barium. Since there are about equal amounts of fission product ^{133}Cs as ^{137}Cs, the cost in neutrons is absolutely prohibitive without isotope separation. According to Ref. 1, it is necessary to separate the ^{137}Cs from ^{133}Cs to a purity of .9999 to reach class A levels for radioactive waste. Equation 9 can be used for an assessment of centrifugation for cesium separation. The parameters used for this evaluation are as follows:

$M_a - M_b = 0.004$ Kg
$\omega = 4397$
Starting concentration of ^{137}Cs, $x_0 = 0.477$
$T = 373$ K.

The practical implementation of centrifugation for Cs is enhanced by two factors; the low melting point of Cs metal of 28.4 °C, and its low viscosity, which we discuss later. The temperature appears in the denominator and enhances the separation significantly. Also the operation at low temperature allows the use of a wider range of high-strength materials making higher speed centrifuges less difficult to build. Therefore this calculation is done only for the higher rotational speed. The volume of Cs which must be processed per year from one 3000 MWt commercial reactor is also small at about 30 liters so that longer residence times are possible and fewer centrifuges are necessary to satisfy the throughput requirements. For this case we take major advantage of feedback as shown in Fig.5 since the separation per stage is low for a non-fedback system. A heavily fedback system is practical since the throughput requirement is low. Starting with a material of concentration $x_0 = 0.477$, the concentration at the inside and outside are found from Eq. 9 to be 0.492 and 0.462 respectively, the change being +- 0.0148. If the material is fed back into the centrifuge with 90 % of the flow coming from the output of the same centrifuge, the composition of the input becomes 0.9 X .492 + 0.1 X .477 = 0.4903. The output enrichment will be .4903 + .0148 = .5051. Upon the second circulation the feed will be 0.9 X .5051 +.1 X .477 = .5023 and the output will be 0.5171. As the material continues to circulate the concentration will increase to a limit, which is estimated by repeated calculations of this type, to be about 0.625.

Fig. 5 shows four centrifuges connected together with the performance estimates indicated. The figures inside the boxes, which represent centrifuges, show the feed fractions at the top and the output fractions at the bottom. At the bottom below the boxes the output fractions of ^{133}Cs are shown. Fig. 6 shows the estimated concentration of ^{137}Cs in the ^{133}Cs output from an array of such interconnected centrifuges. The objective of four nines purity is reached with 12 stages. The separation could be done with fewer centrifuges. For example the output from the first four has a ^{137}Cs composition of about 0.06. This material could be collected and used as a feed material for a second campaign which would improve the composition to 0.0019. This in turn could be used as feed in

a third campaign to reach the a composition of about 0.00005. While the three separate campaigns require fewer centrifuges, the advantages of a continuous closed flow system are considerable and the most appropriate arrangement would require a careful analysis of many factors.

This analysis has neglected the ^{135}Cs which is present in the commercial reactor waste at an isotopic fraction of about 0.12. The concentration of ^{135}Cs in the ^{133}Cs will be greater since the mass difference is two rather than four, so that the concentration would be approximately $(.00005)^{1/2} = .0071$ if it were in the same abundance as the ^{137}Cs. Since there is only about 30 % as much ^{135}Cs as ^{137}Cs to begin with, the abundance of the ^{135}Cs in the ^{133}Cs output would be approximately 0.0021. According to reference 1 this meets the limits for Class A nuclear waste.

The estimates of centrifuge performance presented here are exceedingly attractive and would make the transmutation of ^{137}Cs and ^{135}Cs quite feasible from a neutron economy point of view. A more detailed analysis of the isotopic separation prospect appears to be warranted.

VI. Centrifugal Removal of Lead Spallation Product

The build-up of spallation product in the liquid lead target is a potential problem since eventually it might reach a level beyond which it is not soluble in the lead. The distribution of the spallation products[9] as a function of atomic number (Z) is shown in Fig. 7. Most of the spallation product is close to lead in atomic number and it is nearly as effective as lead as a spallation target. This material also moves back towards lead in mass over a long period because of successive neutron capture. If the lead is in use long enough, this capture process will be balanced by the spallation product production rate. If this equilibrium condition is reached at concentrations of heavy spallation product which are soluble in the lead, then the heavy spallation product need never be removed. The elements in this group include Bi, Tl, Hg, Au, Pt, Ir, Os, Rh, W, Ta, and Hf; many of these are noble metals.

There are many lighter spallation products which are made in very small quantities also. These probably will never work their way back up to lead because too many neutron captures are required. It might be desirable to remove them. These nuclei extend down even to the lightest elements, but the number lower in atomic number Z than 25 is very small. The densities of the elements as a function of Z are shown in Fig. 8. The average density of these lighter materials is about 7.0 compared to 11.35 for lead so that the centrifugal separation should be quite effective. It should also be pointed out that the heavier and more abundant spallation products listed above all have densities substantially higher than lead. Therefore, these will move to the outer radius under centrifugation, and the lighter materials should move to the inner radius with the greatest concentration of lead in between. The concentration of the lighter elements will always be very dilute; the objective in centrifugation would be to concentrate them to a mole fraction of about 0.10 in the centrifuge before

removal. The lead can then be distilled away to give pure spallation product which can either be stored or transmuted to stable material if necessary in the blanket.

The condition of a very dilute lighter component in the centrifuge has not been considered and we examine this case by returning to Eq. 8. We take $x_0 << 1$, which is the case also for $x(r_1)$ and $x(r_2)$. Eq. 8 then becomes

$$[x(r_1) - x(r_2)]/ x_0 = V_a(\rho_b - \rho_a)\omega^2(r_2^2 - r_1^2)/2RT \qquad (11)$$

where ρ_b and ρ_a are the densities of the lead and the light spallation product respectively and V_a is the molar volume of the light spallation product. The following parameters are used to evaluate the effectiveness of centrifugation for lead cleansing: $\rho_b = 11.35$, $\rho_a = 7.0$, $V_a = 17\ cm^3$, $x_0 = 0.01$, $T = 673$ K, $\omega = 2827/s$, $r_2 = 15$ cm, and $r_1 = 5$ cm. We find $(x_1 - x_2)/x_0 = 1.062$, which may be rewritten, $(x_1 - x_2)/2 = 0.531\ x_0$. Therefore the first stage of centrifugation would lead to an increase in the concentration of the light product from .01 to .01531 at the inner radius. Using the concentration 0.01531 as the feed for a second stage gives a concentration at the inner radius of 0.0234. The concentration for six stages is as follows:

Stage	Concentration (lighter spallation product)
0	0.01 (feed)
1	0.0153
2	0.0234
3	0.0358
4	0.0548
5	0.0838
6	0.128

Starting with the third stage the material from the outer radius can be fed back into the first stage and so on with an enhancement of the final concentration beyond the 0.128 value for stage 6. If half of the product is removed from the inner radius at each stage, 1/4 of the material fed into the centrifuge array emerges from the final stage.

The heavier elements with an average density of 20 will be concentrated at the outer radius. Using the parameters $\rho_a = 11.35$, $\rho_b = 20$, $V_a = 18.3\ cm^3$, $x_0 = 0.01$ and all of the other parameters the same, we find the concentration at the various stages to be:

Stage	Concentration (heavier spallation product)
0	0.01 (feed)
1	0.0226
2	0.0511
3	0.116

A concentration of the heavy spallation product near 0.10 is built up far sooner than that for the light spallation product. If removal of both light and heavy spallation product appears to be advisable, the heavy product could be removed using a bank of three centrifuges whenever the heavy product concentration reached 0.01. The light spallation product would require removal much less frequently. The removal of the light product could be done by using the same array of three centrifuges applied to the heavy product separations and cycling twice through the set of three.

VII. Separation Rates

The separations derived in 8 and 9 are equilibrium separations. Whether or not these separations are of more than academic interest depends on the separation rates. Rates are estimated here for the cases considered above. Since the material must be separated at adequate rates to be useful, these rate estimates make it possible to estimate the number of centrifuges of a given size that must be used.

The discussion of rates is simplified if one asks for the drift rate of an isolated molecule of one species through a second material. In a centrifugal field an atom experiences a force $F = M r\omega^2$ which causes it first to accelerate and then to reach an equilibrium velocity depending on the frictional force which according to the Stokes formula is proportional to velocity. This friction force is found to be $F = 6\pi a\eta v$ where v is the drift velocity, η is the solution viscosity, and a is the molecular radius of the moving molecule. Setting these forces equal gives for the drift velocity

$$v = M\omega^2 r/6\pi\eta a \qquad (10)$$

We first apply this to the separation of the carrier from the fission product and actinide ("the "heavies"). Since the carrier is almost all LiF, we use that viscosity for t carrier. This viscosity is not known to the author, but we take it to be somew t larger than the value at 600 °C for LiCl, which is 1.4 centipoise, and assume the value of 2 centipoise for the carrier. We estimate the velocity of a UF_4 molecule in the carrier. The mass is 314 grams/mole and the molar volume assuming a density of 6.7 is 47 cm^3. Assuming 6×10^{23} molecules/mole and the atom volume $V = 4\pi a^3/3$ gives a radius $a = 2.7 \times 10^{-8}$ cm. Taking an average radius in the centrifuge of 10 cm and a value for $\omega = 2827/s$, we calculate a velocity of 4.1×10^{-6} cm/s. Since the average distance which a molecule must move in the centrifuge is (15-5)/2 = 5 cm, the time required for separation is about 12 days. For a volume in the centrifuge of 125 liters and a molten salt volume of 1600 liters, a single centrifuge could process the carrier through the first stage in (1600/125)X 12 = 153 days.

If we require that the first stage separation be repeated once per month, five centrifuges will be required in the first stage. The second stage will require half as many since only half of the material is sent forward to the second stage

The third stage will require only half again for the same reason. The total number of centrifuges for eight stages of carrier separation is then 5 + 3 + 2 + 1 +1 +1 +1 +1 = 15 If the higher speed of 4397/s is used the force is larger by a factor of about 2.5 so the number of centrifuges for the five stages required at the higher speed becomes 2 + 1 +1 +1 +1 = 6 stages. The separation for the fission product will be somewhat slower since the mass is smaller even though the molecular radius is also somewhat smaller. Nevertheless, these estimates place an approximate scale on the size of the centrifuge assembly required for the carrier separation.

For the actinide-fission product separation, the rates are assumed to be about the same. However the volume of the heavies is smaller by about 50 so that only one centrifuge must be used for each stage. A smaller gap (r_2 = 15 and r_1 = 10 cm) in the centrifuge also can be used without sacrificing much in separation performance. The residence time is then 6 days in each of eight centrifuges for the slower rotational speed and 2.5 days in each of the four stages for the higher speed. The total residence time in the centrifuge for actinide-fission product separation is then 6 X 8 = 48 days and 2.5 X 4 = 8 days for the two respective speeds.

The total time for a molecule to pass through both the carrier separation and the actinide removal process is then 15 X 12 + 48 = 228 days for the slower speed and 5 X 5 + 8 = 33 days for the faster speed. In the case of the slower speed the actinide molecule spends 62 % of its time outside the transmuter blanket so the effective flux on the actinide is reduced by the factor 0.38. For the faster speed, the fraction of the time spent outside is 33/ 365 = 0.0904 and the effective flux is reduced by the factor of 0.91.

The gain from increasing the speed by a factor of 1.55 is quite significant. The total amount of the material which must be handled at any one time outside the system is reduced by a factor of 2.5, the number of centrifuges is reduced by the same factor, and the effective flux is increased by the same factor. If the capital and operations cost for the separation is proportional to the number of centrifuges, then these costs are also reduced by the same factor. For a capital cost of the centrifuges of $250,000 each, the total capital cost of the centrifugation system for the slower speed system might be about $6 million. There is a high premium on achieving as high a speed as possible while keeping other parameters the same.

For the isotopic separation of cesium, the drift rates are substantially higher. The viscosity of the liquid cesium at 216 °C is about 0.3 centipoises[8] so that the drift rate is found to be about 5 cm in 1.5 days. However the ^{133}Cs atom must go through 12 stages and is recycled about 20 times in each so that the total time spent is about 12 X 20 X 1.5 = 360 days. The throughput per year for the 12 centrifuges is then 12 X 125 liters X 1.88 = 2820 kg. The annual processing requirement for a 3000 MWt system is about 77 kg, so that the throughput is far too high. The 5-15 cm radius system is not scaled properly for cesium separation. A smaller radius centrifuge with lower volume capacity and higher rotational speed would probably be much more effective. Perhaps r_2 =

7.5 cm, r_1 = 5 cm , and l = 50 cm corresponding to a volume of 5 liters is about right.

For the separations associated with the lead target, we use the viscosity of lead of 2.3 centipoise[6] for a temperature of 673 K and calculate an average atomic radius of the lighter spallation product of 1.9×10^{-8} cm assuming ρ = 7 and A = 120 and for the heavy spallation product 1.56×10^{-8} cm assuming ρ = 20 and A = 190. The drift velocities for these cases are found from 10 to be 1.94×10^{-6} cm/s and 3.74×10^{-6} cm/s respectively.

For the heavy product the residence time is about 13 days per centrifuge. For a volume of 125 liters per centrifuge and a total lead volume of 1500 liters, the processing time for all of the lead would be 156 days. The time spent by a single atom in passing through the centrifuge would be 39 days. By using two centrifuges for the first stage and single centrifuges for the following two stages, the capacity of the centrifugation would be doubled and the processing time would be reduced to 78 days. One such campaign might be run annually.

For the light spallation product separation, the velocity is slower and the number of stages required is larger. The separation time per centrifuge is about 26 days. For the set of four centrifuges described above (three centrifugation stages), 250 liters could be processed in 26 days; the total of 1500 liters could be processed in 156 days. The volume of lead would be reduced by four for this first cycle through the three stages. Therefore the second cycle through could be completed in another 50 days for a total of 206 days . Since it takes a single atom 78 days to pass through three stages, these times would be extended somewhat. It seems likely that removal of the light spallation products might require a campaign lasting about one year. One such campaign might be run perhaps every ten years. Of course, the use of the higher speed centrifuge would reduce the number of stages and the processing time substantially.

The frequency of removal will depend ultimately on the total volume of lead in the target, the proton beam current, the solubility of the spallation products, and the neutron capture rate in the spallation product. The capture rate in the spallation product should be kept small compared to that in the lead. In summary, it appears that centrifugation might be an effective and low cost means for dealing with spallation product removal from the lead target. The absence of a waste stream associated with this process, except for that in maintenance of centrifuge components, is an attractive feature of this approach.

VIII. Non-Ideal Solutions

The analysis described above was done for an ideal solution. For a binary system an ideal solution exists when the forces between molecules are not necessarily zero but when the forces A-A, B-B, and A-B are the same. If these forces are not the same, then the solution will be non-ideal and energy may be released or absorbed in changing the concentration by some means

such as centrifugation. Therefore additional energy may be necessary beyond that required to overcome the entropy of mixing. One way of looking at this question is to compare the centrifugal force with the "entropy force." For a binary solution with a concentration gradient, the search for maximum entropy will drive the solution to a uniform concentration. The "phantom", "effective", thermodynamic force is $F = -d\mu/dr$ where μ is the chemical potential per mole of mixture. This force is zero when the mixture is uniform. It attempts to push the mixture to uniformity when another force such as centrifugation upsets the uniformity or when two liquids are poured together. In an ideal binary solution, the Gibbs free energy per mole m is given by $\mu = \mu_0 + RT\ln \gamma x$ where x is the concentration of component a in the mixture of a and b and γ is a number between 0 and 1 which measures the degree of departure from an ideal solution. It is concentration dependent and therefore depends on r.

We consider first the case for an ideal solution for which $\gamma = 1$. Therefore this force of mixing per mole of solution is $F = -(RT/x)(dx/dr)$. In a mole of solution there are xA molecules, where A is Avogadro's number, so that the force on a single molecule is $F_{mixing} = -(RT/x^2A)(dxdr)$. The centrifugal force on an individual molecule of solute is $-mr\omega^2$ where m is the mass of an individual molecule. The mass m can be related to the molecular weight M by the relationship $m = M/A$ so that the centrifugal force per molecule $F_{centrifugal} = -Mr\omega^2/A$. The ratio of these forces is then

$$F_{mixing}/F_{centrifugal} = RT(dx/dr)/x^2Mr\omega^2. \qquad 11$$

We see that for very dilute solutes with $x << 1$, the centrifugal separation might be rather unfavorable since the mixing force is strongly enhanced by low concentrations. If we apply 11 to the separation of carner from the heavies, we know from Figs. 2 and 3 that for an initial concentration of 0.014 for the heavies, the separation achieved over a distance of 10 cm was 0.994 - 0.978 = 0.016 so that in MKS units $dx/xr = .016/0.1 = 0.16\ m^{-1}$. Using M = .248 Kg, $\omega = 2827$, r = 0.1 m, and T = 873 K gives $F_{mixing}/F_{centrifugal} = 29.9$. Therefore the centrifugal force is weak compared to the mixing force and only a small degree of separation is possible. However, in subsequent stages, the value of x becomes larger and the degree of separation with each stage rapidly becomes larger because of the strong dependence on x, as can be seen from Fig. 2.

Consider now the case for a non-ideal mixture for which $\gamma < 1$. The force F_{mixing} is

$$F_{mixing} = -(RT/A)(dx/dr)[(\gamma/x^2) + (1/\gamma)d\gamma/dx] \qquad 12$$

We see from the first term on the right hand side of 12 that the force is reduced by the value of γ from the force for an ideal solution. The second term in the force is not dependent explicitly on the molar concentration x but only on the variation of γ with r. For a very dilute solute, the value of $\gamma = 1$ and it doesn't

change much for $0 < x < 0.15$. The reason for this is that at $x < .15$ the molecule is surrounded on all sides by six neighbors of the other species in the solution and making the solution more dilute does not change this. Therefore for $x < 0.15$, γ is very near unity and depends only very weakly on x. Referring back to Fig 2, any non-ideality of the solution has very little effect until the sixth stage of separation. Beyond the sixth stage, some effects on the separation might become observable. The degree of the influence of $d\gamma/dx$ can only be determined by measurements of the dependence of γ on concentration in the concentration range of interest ($0.8 > x > 0.65$). In concluding, it is important to note that while including the non-ideality of the solution will change the mixing force, it will not eliminate the separation by centrifugation. It will only change the degree of separation per stage.

The effect of non-ideality on the actinide-fission product separation could be more significant since the beginning concentration is close to $x = 0.5$ and the dependence of g on x might be significant. However, this is not a binary mixture but a mixture of at least 30 different fluoride salts. Therefore any individual molecule will not in general be surrounded by six molecules of the same species or by a molecule of a single different species. Rather it usually will be surrounded by molecules of six different species. It appears that the solution therefore might approach ideal conditions by virtue of its complex composition alone. As the separation proceeds to greater purity or removal of actinide, the actinide molar concentration becomes small. In contrast to the situation for carrier removal where conditions change from an ideal to a non-ideal condition, in this case the system starts perhaps at a non-ideal condition and moves towards an ideal condition as the actinide concentration is decreased. It seems unlikely that this multicomponent mixture of high order would manifest significant non-ideality at any actinide concentration. The effect of any non-ideality would be to reduce the separation per stage....not to eliminate the separability by centrifugation. However, it is highly desirable to demonstrate the expected performance by actual centrifuge experiments.

For the case of isotopic separation, all of the molecules are of the same chemical species so that there can be no dependence of γ on x and there are no uncertainties introduced by concerns about non-ideality.

For the case of lead separations, the spallation product is always at small concentration as long as the spallation product is not allowed to accumulate or be concentrated to molar concentrations larger than 0.15 so that non-ideality is probably never an issue for the centrifugation associated with the lead target.

IX. Summary

An equation has been derived for assessing the prospects for applying liquid centrifugation to separations processes important to the accelerator transmutation of nuclear waste. It can be shown from this equation that limitations on the centrifuge geometry and speed imposed by available materials limit the degree of separation per stage to the order of a few percent.

Nevertheless highly useful separations can be achieved by making use of several stages of centrifugation. The actinide and fission product is present in the molten salt carrier at concentrations near 1 % and centrifugation has been shown to be promising for increasing this concentration to the 30 % level. Centrifugation has been shown to be promising in providing a fission product stream for further separations which has a quite low actinide contamination. The isotopic separation of Cs in a heavily fed-back system might make practical the possibility of transmutation of the ^{137}Cs and ^{135}Cs in commercial nuclear waste. Also it might be possible to cleanse the spallation product from the lead target separating both the high density and the low density spallation product in separate campaigns.

The drift speeds of the molecules being separated has been estimated and these speeds used to estimate, in turn, the number of centrifuges required and the practical times required for separations. It has been shown that the introduction of improved materials with high strength at high temperatures, and low density can have a very large effect on the performance of these centrifugation concepts. The basic concept of centrifugation makes possible a closed separations system without the use of reagents, etc. and the necessity to deal with their associated waste streams. The systems are well adapted to unattended operation and hands-off maintenance and control. Since the separations are performed on liquid salts or metals, there is no effect of radiation on the performance of the centrifugation except perhaps in the temperature control. The effects of radiation-induced organic material damage and the uncertainties of ionization on aqueous chemical separations need not be of concern. Therefore the expected performance can be verified with high reliability in laboratory centrifugation experiments without the use of radioactive material, except for the case of monoisotopic cesium.

There are a number of extensions of this analytical assessment which should be pursued:

1. The assumption of equivalence of partial molar volume and molar volume should be examined.

2. The cases where departures from an ideal solution are significant should be investigated.

3. The technique should be extended to multicomponent systems where appropriate.

4. The drift speeds can be made more quantitative.[3] The radial drift rate will not be constant with radius, but will be influenced by the radial dependence of the centrifugal field and by the approach to equilibrium.

5. The staging and interconnection of the centrifuges together must be investigated and optimized making use of the exact solutions of the appropriate differential or difference equations.

6. The flow of the liquid through the centrifuges must be examined carefully since that might impact performance. For example, the axial flow velocity of the salt is not uniform with radius but parabolic with the flow rate at the wall being zero. Furthermore care must be taken in extracting the separated components to extract near the inner or outer wall. Drawing material from the center will compromise the separation performance of the system.

7. The use of centrifugation to concentrate the components of a solution suggests the possibility for inducing precipitation as a result of the centrifugal concentration. Enhanced separation would be associated with this precipitation-centrifugation combination.

In conclusion, this study suggests that centrifugation might address well some of the most difficult challenges for radiochemistry posed by transmutation and partitioning technology. The study here does not eliminate the need for aqueous chemistry, but it very substantially reduces the aqueous chemistry required and provides an effective means by which aqueous chemistry can interface with molten salt and perhaps other pyrochemical processing.

Acknowledgements

The effort of R. J. Jensen in performing scoping calculations suggesting that centrifugation, among other possible physical-chemistry methods for ATW, was worth further study is much appreciated. His encouragement of W. D. Breshears and J. L. Lyman to push forward on this subject was an important step since it provides independent means by which the Laboratory can verify the fundamental principles of centrifugation for the partitioning required by ATW. Comments of A. G. Petschek on the non-uniform axial flow, the need for care in the extraction of the separated components at the inner and outer radius to avoid a flow-related compromise in performance, and on the non-uniformity of the radial drift rate are much appreciated.

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Physical properties and operating limits of possible centrifuge materials

Material	Aluminum alloys	High-tensile steel	Titanium	Maraging steel	Glass fiber	Carbon fiber/ resin
Density						
g/cm^3 †	2.8	7.8	4.6	7.8	1.8	1.6
kg/m^3 (ρ)	2,800	7,800	4,600	7,800	1,800	1,600
Tensile strength						
kg/cm^2 †	4,570	14,080	9,150	19,700	5,000	8,450
MPa (10^{-6} σ)	448	1,381	897	1,932	490	829
Modulus of elasticity						
Mg/cm^2	724	2,110	1,160	2,110	738	
MPa (10^{-6} E)	71,000	207,000	114,000	207,000	72,400	
Max. tangential speed, $v_{max} = \sqrt{\sigma/\rho}$, m/s	400	421	442	498	522	720
Length-to-radius ratio at v_{max}, Eq. (14.153)						
First resonance	14.0	13.8	13.2	13.8	13.8	
Second resonance	23.4	23.1	22.2	23.1	23.0	
Third resonance	32.8	32.4	31.1	32.4	32.2	
Fourth resonance	42.2	41.6	39.9	41.6	41.4	
Fifth resonance	51.5	50.8	48.8	50.8	50.6	

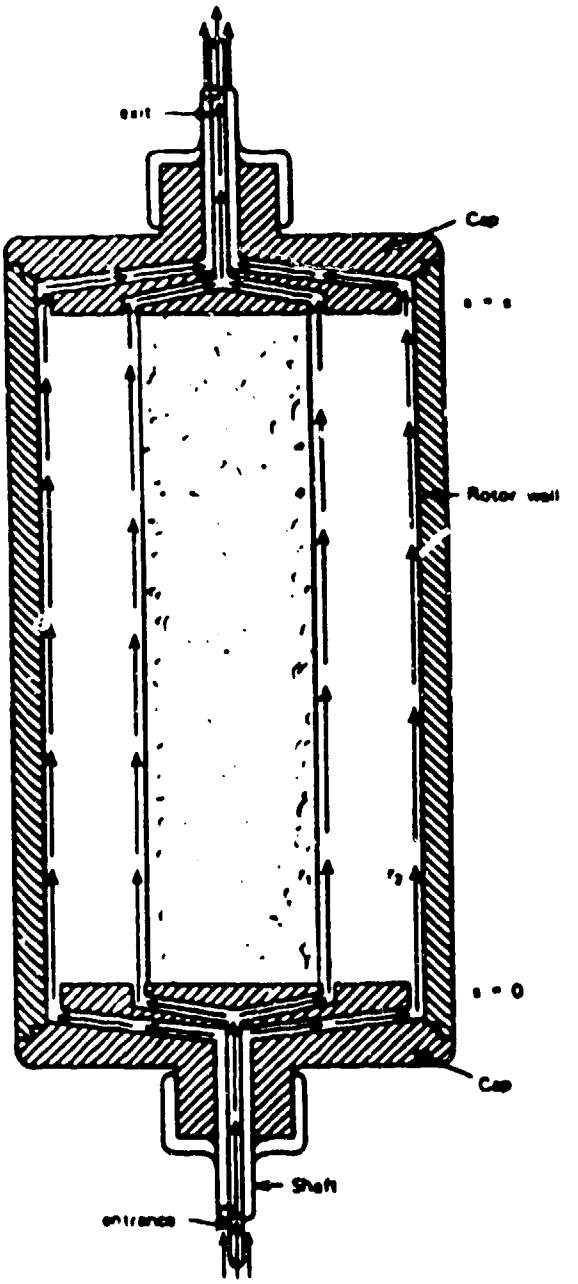

Fig 1 Schematic diagram of a simple centrifuge with two entry points at the bottom and to exit points at the top. For the centrifugation discussed here, the material enters through only one port at the bottom. It leaves at both the inner and outer radius with the ratio of exit flows determined by external valving not shown The centrifuge is usually completely full of solution.

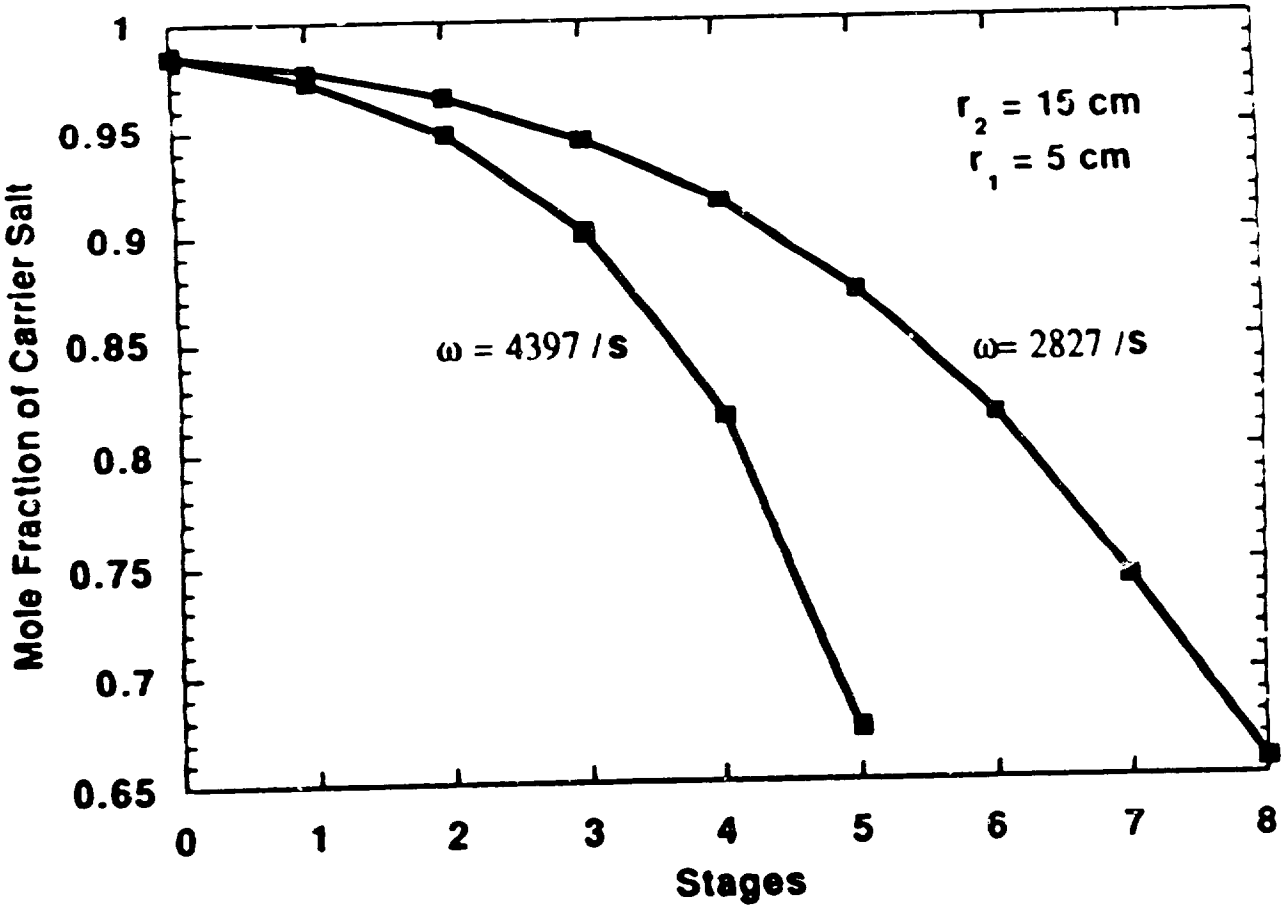

Fig. 2 The separation of carrier salt ($LiF\text{-}BeF_2$) from the heavier actinides and fission products. The centrifuge has inner and outer radii of 5 and 15 cm respectively and is assumed to spin at an angular speed of 2827 or 4397 radians per second. The concentration is shown as a function of the stage. The salt enters as 0.986 mole fraction carrier and the concentration dependence on the number of stages in the array is shown. At a mole fraction of 0.65, the volume fraction of the carrier is actually about 0.31.

ATW Carrier Salt Separation

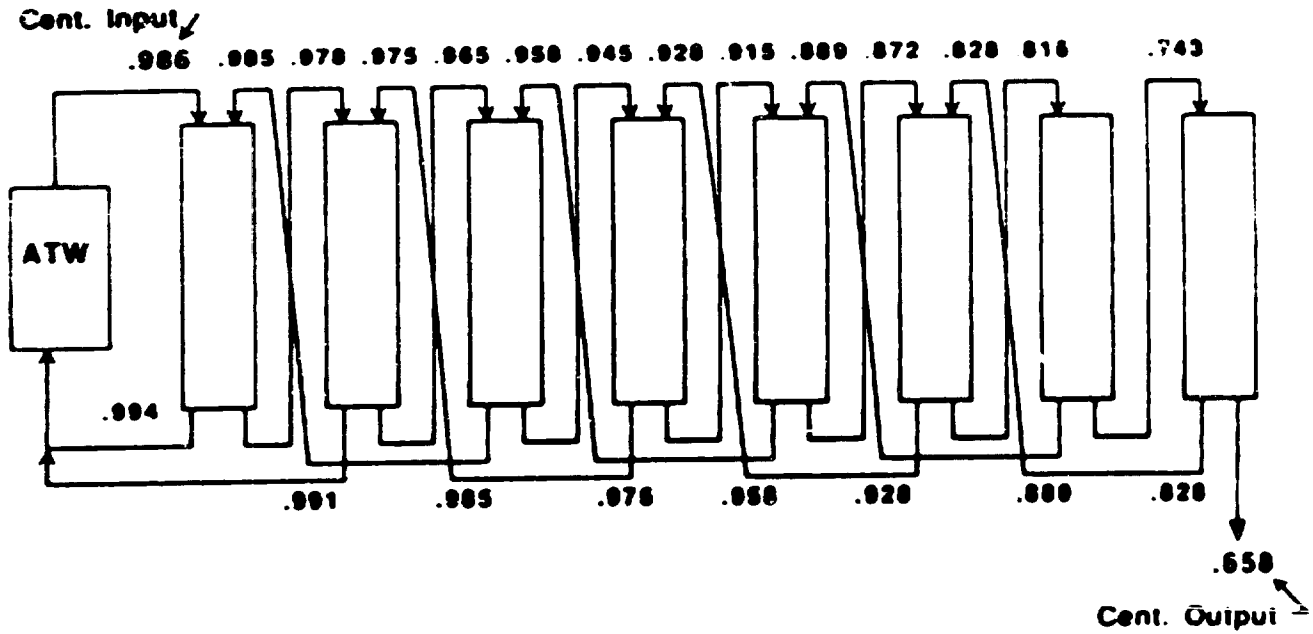

Fig. 3 The eight-stage separation of carrier salt. The stage interconnections and the concentration at each connection is shown for a speed of 2827 radians/s. The material enters from the blanket at a mole concentration of 0.986 carrier. The salt removed from the inner radius has a concentration of 0.994 and is returned to the ATW blanket. The concentration of the carrier in the outer radius is 0.978 and this is sent on to the second centrifuge. The carrier concentration from the inner radius of the third centrifuge is 0.985 which is a slightly lower mole percent than the feed from the ATW, so it is recycled into the first centrifuge rather than being returned to the ATW. All of the subsequent centrifuges are similarly recycled.

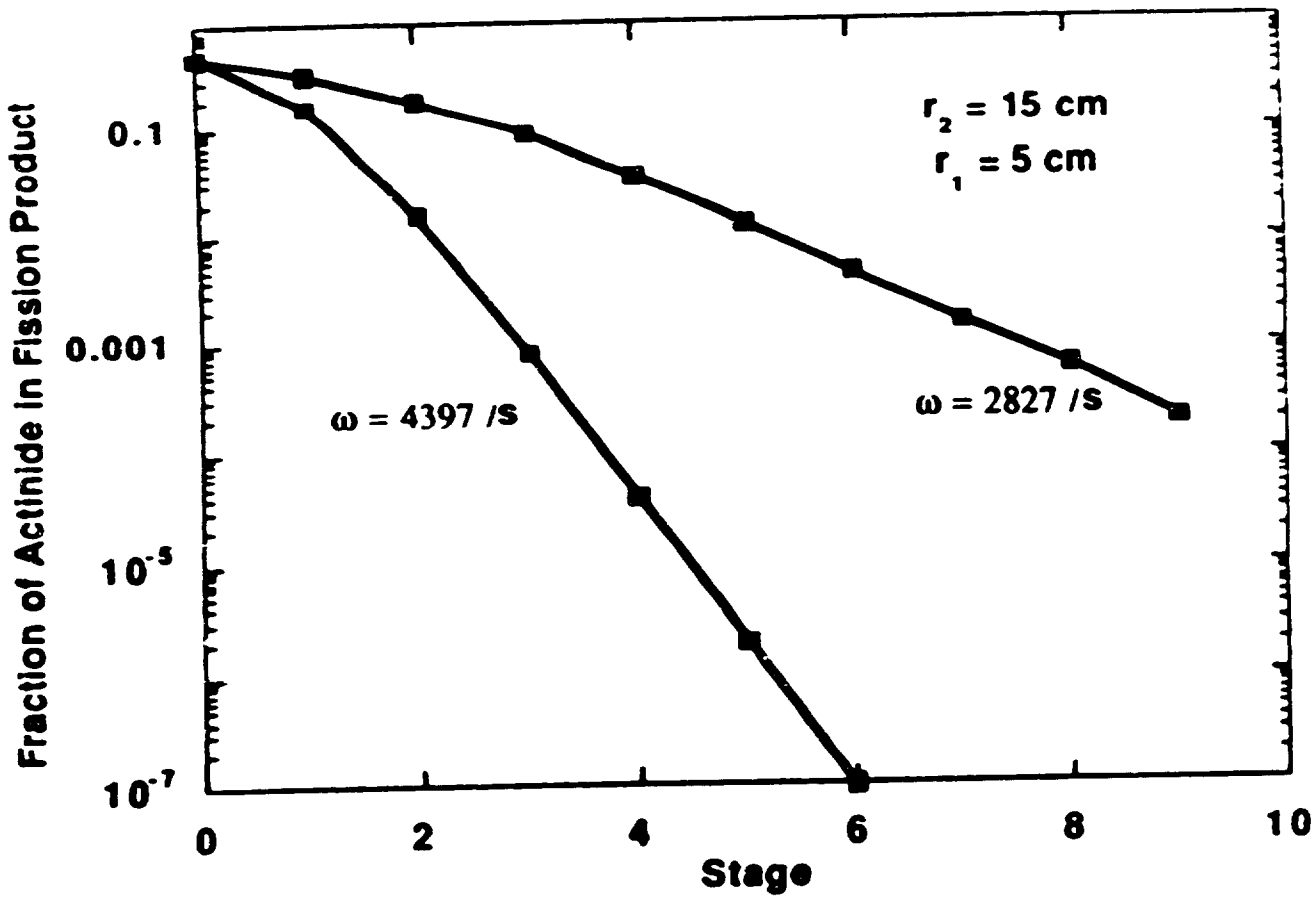

Fig. 4 The Separation of actinide and fission product. The fission product mole fraction feed is initially 0.50 and the concentration dependence on the stage is shown for two angular velocities. Note that the higher angular speed (a factor of 1.55 larger than the other) performs with much greater effectiveness

Cesium Isotope Separation

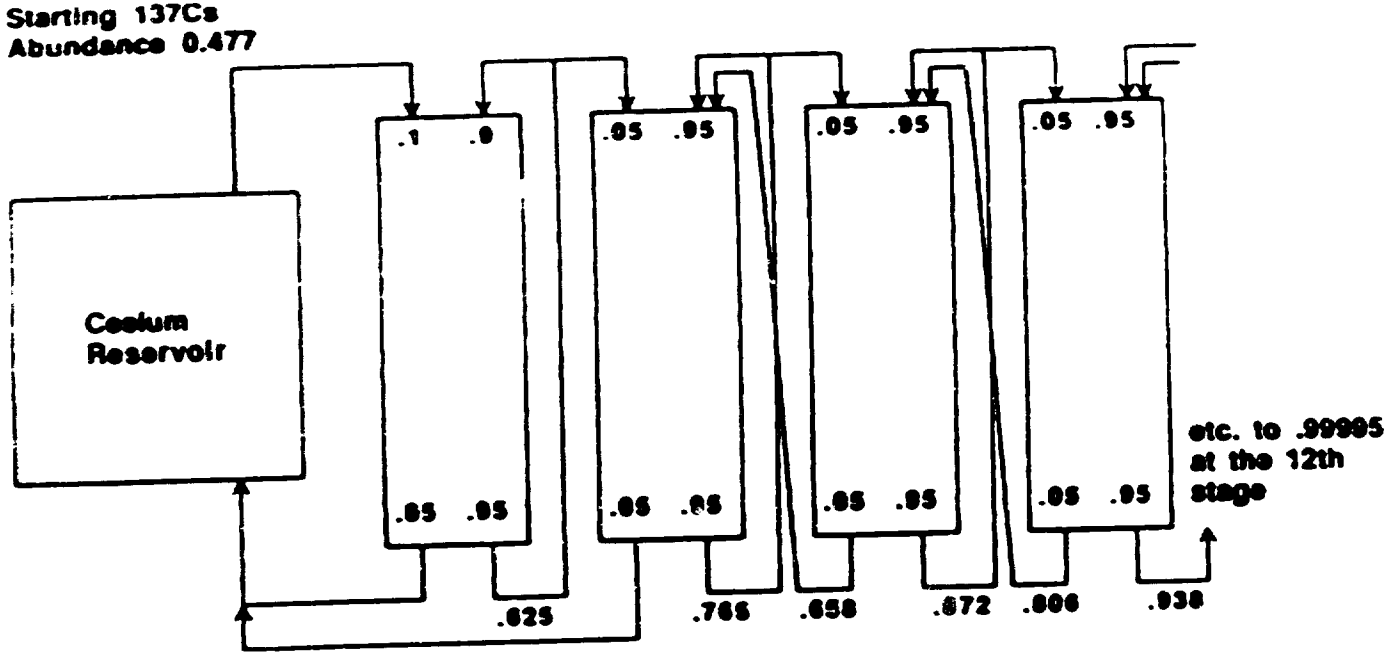

Fig. 5 The centrifuge arrangement for isotopic separation of cesium. Cesium enters with an isotopic abundance of ^{133}Cs of 0.477. The figures at the top of the centrifuges shows the feed fractions for a heavily fed-back system. The figures inside the centrifuges at the bottom show the exit fractions. The third centrifuge receives material from the outer radius of the second; 95 % of the flow is from the outer radius. Ninety percent of the flow from the second centrifuge is returned to the input of the second stage and 5 % is directed into the third stage. The five percent of the flow from the outer radius of the third centrifuge has a concentration of 0.658 which is better than the feed for the second stage, so it returns to the second stage with advantage instead of being returned to the reservoir. By the 12th stage the concentration of ^{137}Cs in ^{133}Cs has been reduced to 0.99995.

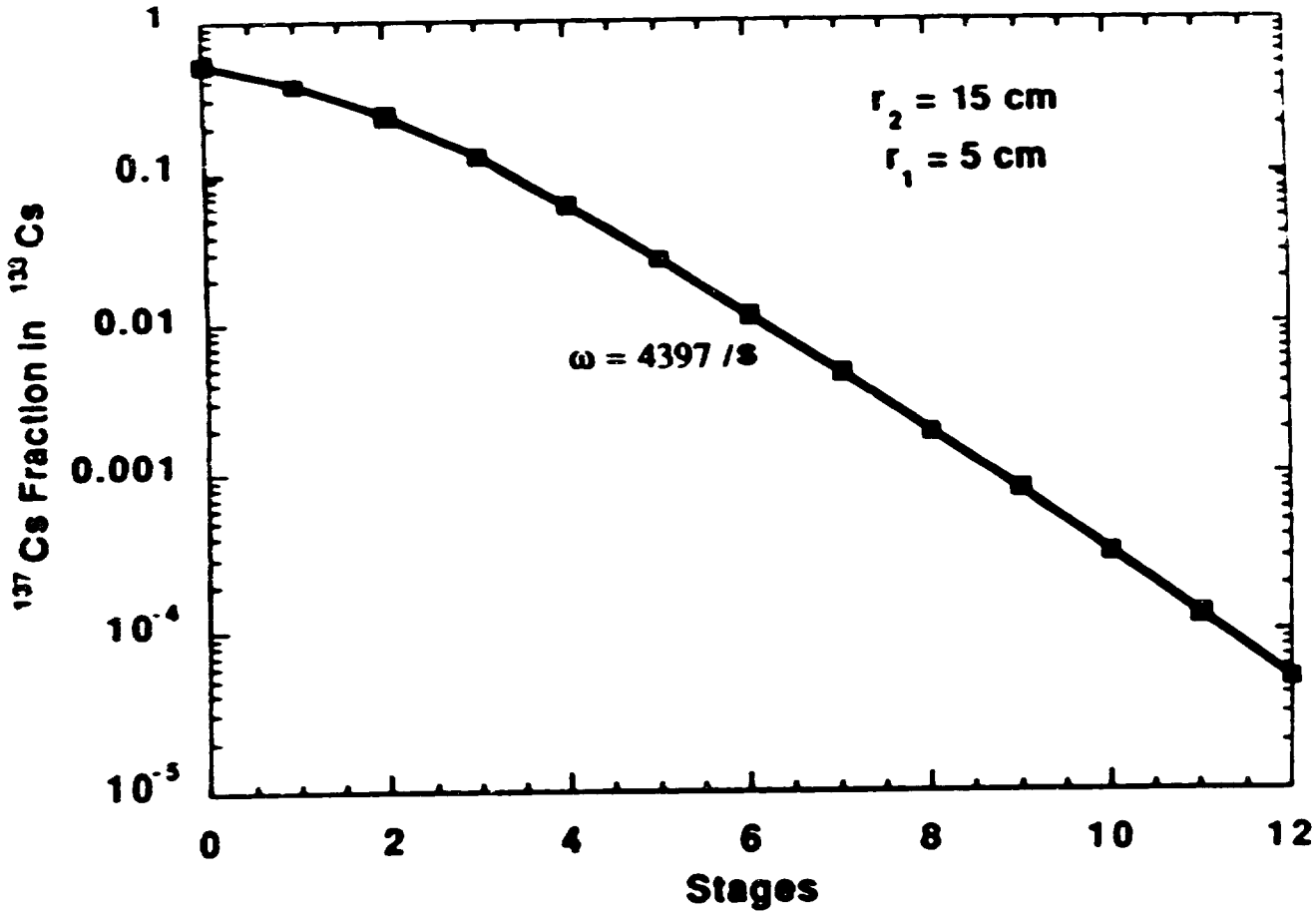

Fig. 6 The fraction of ^{137}Cs in ^{133}Cs as a function of the stage for an initial concentration of 0.477 of ^{137}Cs.

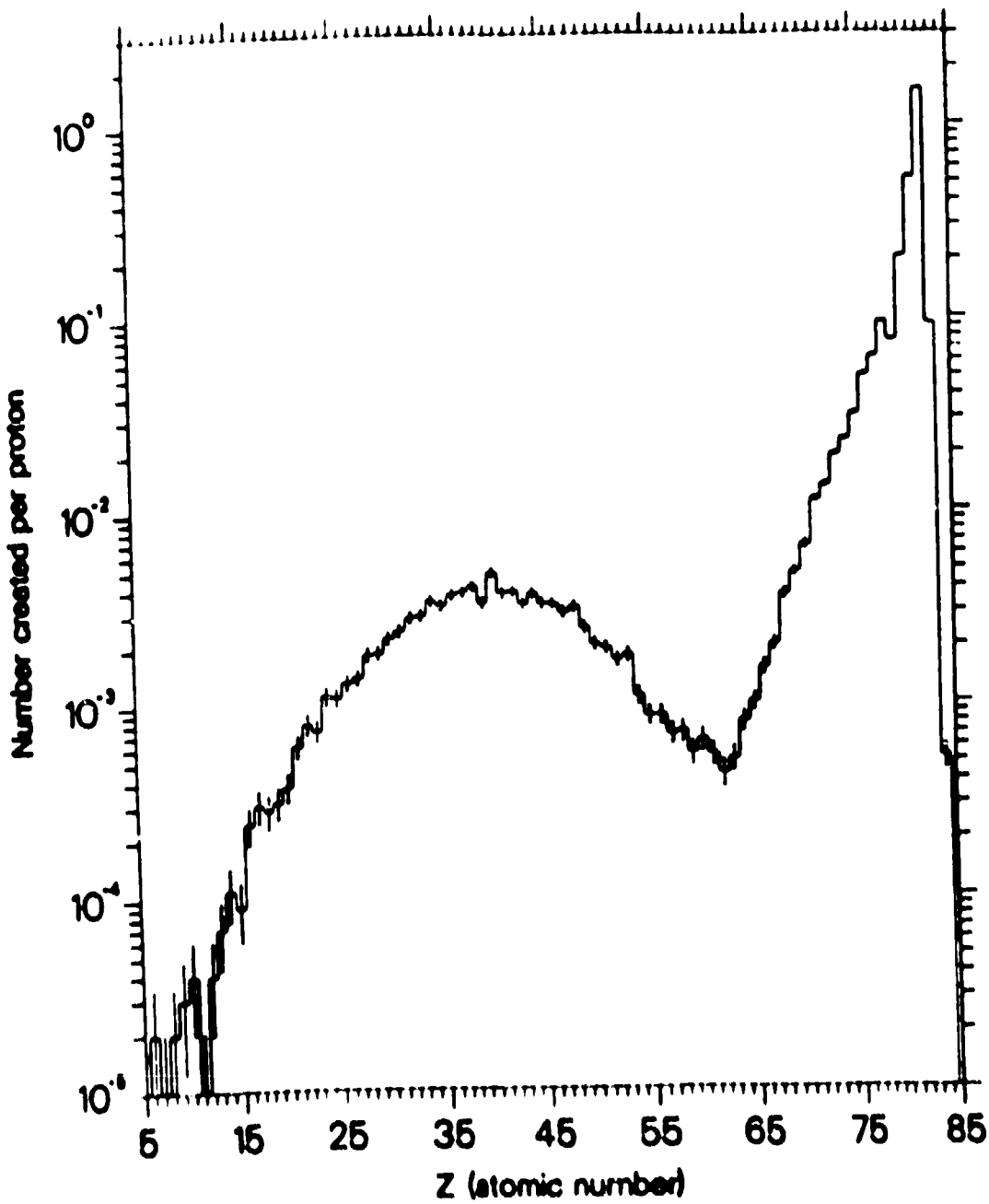

Fig. 7 The spallation product distribution for a lead target bombarded by 800-MeV protons. The figure shows the number of atoms of a particular element per proton as a function of the atomic number. This is the initial Z distribution. The distribution will be changed somewhat by beta decay and by neutron capture.

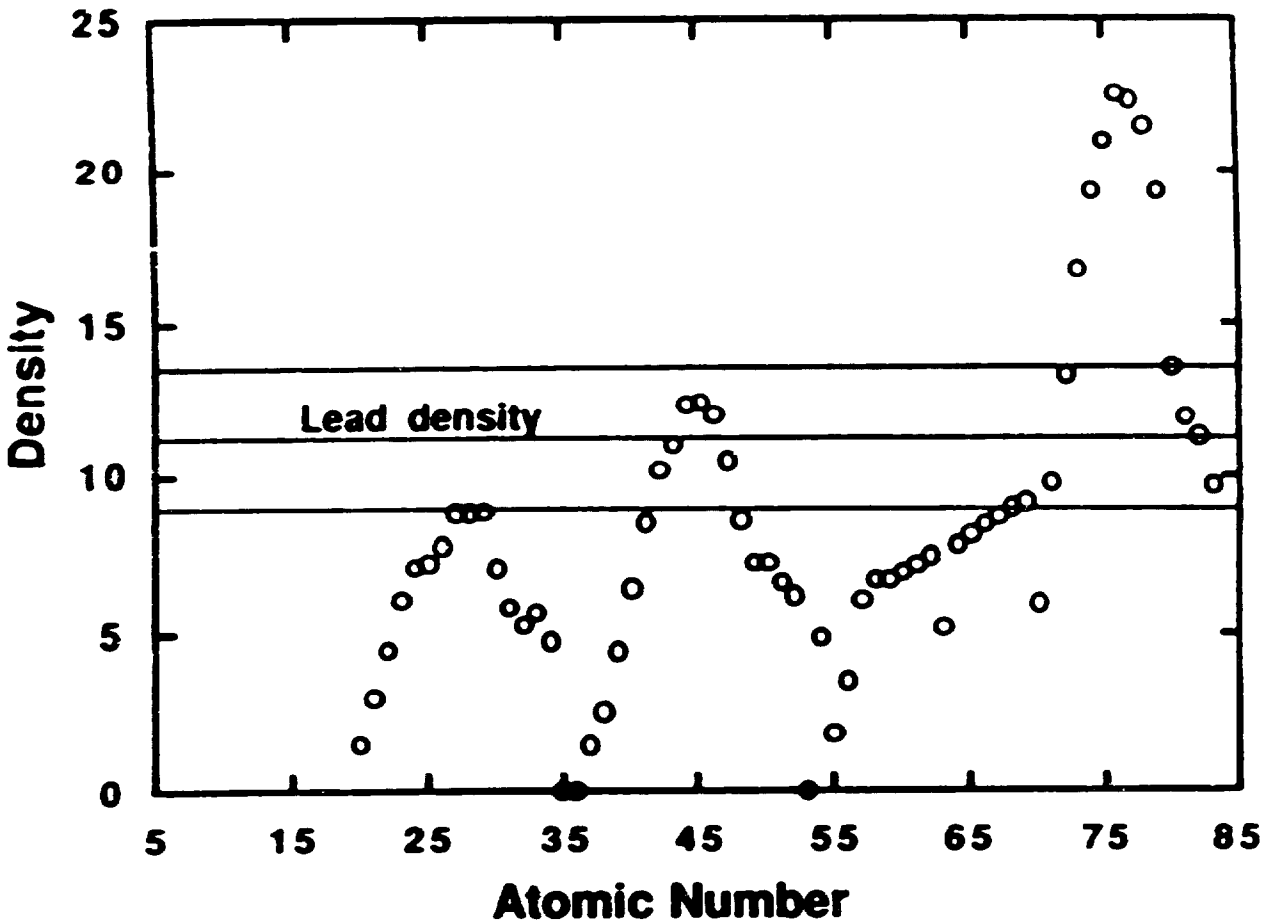

Fig. 8 The density of the elements as a function of atomic number. The horizontal lines around the lead density divide the elements into three groups. The elements in the peak in density near Z = 77 are the most abundantly produced in spallation and will move to the outside under centrifugation. The elements below a density of about 8.5 will move to the inside. The elements in the band around the lead density will not separate easily from lead under centrifugation. They can be removed following transmutation by successive neutron capture to less dense higher Z elements.